5.0 MASS BALANCE ACTIVITIES

5.1 ESTIMATED OVERALL MASS BALANCE OF RECYCLED URANIUM

This chapter combines the gross quantities of enriched RU shipped to/from the Y-12 Complex (Chapter 3) with the constituent analytical data (Chapter 4) and process engineering judgment (Chapter 2) to provide an estimate of the actual quantities of RU and the constituents of concern that passed through the Y-12 Complex. These mass flows are presented in a framework that balances quantities received at the plant against quantities shipped, quantities contained in waste, and those remaining in inventory on-site.

As noted in Chapter 2, only RU materials that would have been chemically processed through 9212 or 9206 (only highly enriched RU) are addressed in the analysis of mass balance activities. RU below this assay range that was shipped to the Y-12 Complex was eliminated from further consideration, as discussed in Chapter 2, in accordance with the methodology established for the RU mass balance project.

5.1.1 Uranium

The total flow of highly enriched RU into the Y-12 Complex, as presented in Chapter 3, was determined to be 150.9 MTU, from years 1953 through 1999. This material was received from the Savannah River Site (125.2 MTU) and the Idaho Chemical Processing Plant (25.7 MTU).

The material was received in the form of uranyl nitrate solution, uranium trioxide (UO_3), uranium/aluminum metal, miscellaneous floor sweepings, and casting dross. Quantities received, shipped, and currently in inventory for the Y-12 Complex are summarized in Table 5.1-1.

Table 5.1-1 Summary of Y-12 Complex Highly Enriched RU Mass Flow

RU Received	Quantity (kg)
Savannah River Site (1955-93)	125,161
Idaho Chemical Processing Plant	25,696
	Total 150,857
RU Shipped	
Savannah River Site	120,384
	Total 120,384
Total RU Inventory	
Currently On-Site	13,082
	Total 13,082
Estimated RU Waste	
	Total ~100
Balance of Material	
	Total ~17,300

Most of this material was processed through the 9212 and/or 9206 facilities to produce metal buttons for shipment to SRS where they were used in the fabrication of reactor fuel rods. The process employed at the Y-12 Complex was basically that of nitric acid dissolution, solvent extraction of uranyl nitrate to separate the uranium from aqueous solution through preferential dissolution and leaving impurities behind in a raffinate stream, denitration to uranium oxide, reduction and hydrofluorination to UF_4, and reduction to uranium metal. The raffinate was discharged as a waste directly to the S-3 Ponds until the mid-1980s, when the WETF was constructed. After that time, the raffinate was discharged to the WETF.

The remaining Y-12 Complex on-site RU inventory is primarily in the form of metal buttons. Some unprocessed U-Al metal scrap, casting dross, and floor sweepings also remain and are slated for processing through Nuclear Fuel Services (NFS) through contracts managed under the DOE Central Scrap Management Office for uranium. Residual amounts of RU were buried with process solid waste as unleached oxide.

5.1.2 Plutonium and Neptunium

The availability of site-specific RU data at the Y-12 Complex, beyond that of gross material receipts and shipments, was limited for SRS material and was available only from Idaho National Engineering and Environmental Laboratory for ICPP materials. As a result, the RU mass balance report prepared by the ICPP was used to determine mass quantities of the constituents of concern that were shipped to the Y-12 Complex from that site, while Y-12 Complex data were used for SRS. As is evident in Table 5.1-1, the SRS material constituted the overwhelming portion of RU processed at the Y-12 Complex.

As reported in Chapter 4, data on plutonium and neptunium constituent levels in RU were found in a variety of forms and locations. Files/records were predominantly obtained from individual personal archives (often in boxes from retired employees) and included actual laboratory reports. In addition, C.M. West (retired Y-12 Complex Health Physicist) developed a presentation for the Department of Energy in 1985[1] that spoke in detail to the issues surrounding RU material at the Y-12 Complex, including constituent levels and potential worker exposures. Mr. West had been employed at the Y-12 Complex during most of the years that RU was processed and possessed a working knowledge of the materials and processes, in addition to his radiological expertise.

Most data reported the material alpha ratio and the associated activities (μCi/g U) for Pu, Np, and Th). Where actual laboratory data or laboratory reports were available, they were used as the primary data sources for the respective period of time.

The individual constituent activity in the RU was combined with the constituent specific activity to arrive at the mass fractions (parts per billion – ppb) for Pu and Np.

A representative Pu specific activity, which is dependent upon the isotopic abundances of plutonium in the material, had to be used. Mr. West refers to a value of 2.77×10^{13} dpm/g U, based upon a mixture of 75% ^{238}Pu and 25% ^{239}Pu. Information subsequent to the West presentation[2] suggests that a more representative value for production reactor grade plutonium would be 3.1×10^{13} dpm/g Pu, based on isotopic abundances of 84% ^{238}Pu, 14% ^{239}Pu, and 2% ^{240}Pu. This more recent figure was used for this report. For the ^{237}Np isotope, the value of 1.57×10^{9} dpm/g Np was used.

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[1] West, C. M., “Radioactive Contaminants in Uranium Reactor Returns at the Oak Ridge Y-12 Plant,” 1985.
[2] “Some Early Results Describing Plutonium Contamination of Highly Enriched Uranium (HEU),” correspondence, J. E. Vath, September 8, 1999.

5.1.2.1 Receipts

Data for receipts of Savannah River material were available from a variety of sources for years 1977 through 1986. Those years prior to 1977 and subsequent to 1986 (through 1989) were much more limited in the availability of representative data. As a result, the years 1977 through 1984 were used as a base period for extrapolation to values representative of other periods of time where data was scarce or unavailable. Consequently, this period of time is discussed first, rather than the normal chronological presentation.

The Pu and Np constituent quantities by year from Savannah River are presented in Table 5.1-2. Table 5.1-3 presents the corresponding quantities for RU from ICPP.

1977 through 1986

Data for this timeframe were predominantly in the form of laboratory reports and/or logbook-type records maintained by the operating group(s). The latter were often chronological listings of the individual lab results routinely received and recorded as a normal part of the operation.

Data from the various sources were generally found to be in good agreement, and the West presentation provided similar results. The data typically included the date, material code/description, identification numbers, constituent activities, alpha ratio, beta ratio, and total uranium alpha. Typically, several individual analyses or data sets were available for each year. The parameters of interest from this raw data were averaged to arrive at representative values for each year and subsequently applied to incoming material quantities to determine annual quantities of Pu and Np.

1973 through 1976, and 1987 through 1989

There were no data available at the Y-12 Complex that directly represented these periods of time. A review of available information (including the SRS Mass Balance Report[3]), however, did not suggest or report any changes in operations or processes at SRS that would be expected to result in significant changes in the constituent levels in materials coming to the Y-12 Complex. The PUREX process continued to be employed at SRS until 1992. Consequently, the average values of the Pu (0.004 μCi/g U) and Np (0.016 μCi/g U) activities from the years 1977 through 1984 were applied to each of these years.

1964 through 1972

The C. M. West presentation reported an average alpha ratio for each of nine different intervals during this time period (December 1964 through May of 1972). Additional information related to this data is included in Section 4.4.1.1. The presentation did not, however, report the corresponding constituent activities (μCi/g U) or fraction of the alpha ratio due to each constituent.

To arrive at representative activity levels for Pu and Np, an average alpha ratio was calculated from the West presentation for 1964 through 1972. The average values for Pu and Np activities from the base period were then adjusted by the fraction of the average alpha ratio from each period of time, 0.43 (1964 through 1972) and 0.26 (1977 through 1986).

[3] SRS, *Historical Generation and Flow of Recycled Uranium at the Savannah River Site*, ESH-PEQ-2000-00059.

Table 5.1-2 SRS Receipts and Constituent Quantities by Year

	Period	Receipts kg	Pu			Np			Tc	
			Average (µCi/g U)	ppb	g	Average (µCi/g U)	ppb	g	ppm	g
	1953									
	1954									
	1955	2	0.007	0.48	0.000	0.027	37,474	0.1	114	0.2
	1956									
	1957	3	0.007	0.48	0.000	0.027	37,474	0.1	114	0.3
	1958	18	0.007	0.48	0.000	0.027	37,474	0.7	114	2.1
	1959	149	0.007	0.48	0.000	0.027	37,474	5.6	114	17.0
	1960	6,235	0.007	0.48	0.003	0.027	37,474	233.7	114	710.8
	1961	2,058	0.007	0.48	0.001	0.027	37,474	77.1	114	234.6
	1962	2,397	0.007	0.48	0.001	0.027	37,474	89.8	114	273.3
	1963	6,446	0.007	0.48	0.003	0.027	37,474	241.6	114	734.8
West Data	**1964**	2,978	0.007	0.48	0.001	0.027	37,474	111.6	114	339.5
	1965	3,552	0.007	0.48	0.002	0.027	37,474	133.1	114	404.9
	1966	3,700	0.007	0.48	0.002	0.027	37,474	138.7	114	421.8
	1967	2,502	0.007	0.48	0.001	0.027	37,474	93.8	114	285.2
	1968	2,109	0.007	0.48	0.001	0.027	37,474	79.0	114	240.4
	1969	4,090	0.007	0.48	0.002	0.027	37,474	153.3	114	466.3
	1970	2,060	0.007	0.48	0.001	0.027	37,474	77.2	114	234.8
	1971	3,500	0.007	0.48	0.002	0.027	37,474	131.2	114	399.0
	1972	4,701	0.007	0.48	0.002	0.027	37,474	176.2	114	535.9
	1973	5,070	0.004	0.29	0.001	0.016	22,539	114.3	114	578.0
	1974	4,581	0.004	0.29	0.001	0.016	22,539	103.3	114	522.2
	1975	5,131	0.004	0.29	0.001	0.016	22,539	115.6	114	584.9
	1976	4,312	0.004	0.29	0.001	0.016	22,539	97.2	114	491.6
Base Period	**1977**	4,505	0.014	0.97	0.004	0.035	48,880	220.2	114	513.6
	1978	2,078	0.003	0.18	0.000	0.007	9,108	18.9	114	236.9
	1979	4,576	0.003	0.22	0.001	0.021	29,767	136.2	114	521.7
	1980	1,489	0.001	0.09	0.000	0.006	8,408	12.5	114	169.7
	1981	4,911	0.004	0.30	0.001	0.008	11,695	57.4	114	559.9
	1982	5,719	0.002	0.13	0.001	0.004	5,561	31.8	114	652.0
	1983	6,649	0.002	0.12	0.001	0.006	8,162	54.3	114	758.0
	1984	4,870	0.004	0.30	0.001	0.025	34,603	168.5	114	555.2
	1985	8,243	0.004	0.30	0.002	0.025	34,603	285.2	114	939.7
	1986	5,718	0.004	0.30	0.002	0.025	34,603	197.9	114	651.9
	1987	4,575	0.004	0.29	0.001	0.016	22,539	103.1	114	521.6
	1988	3,095	0.004	0.29	0.001	0.016	22,539	69.8	114	352.8
	1989	79	0.004	0.29	0.000	0.016	22,539	1.8	114	9.0
	1990	67	0.004	0.29	0.000	0.016	22,539	1.5	114	7.6
	1991									
	1992	272	0.004	0.29	0.000	0.016	22,539	6.1	114	31.0
	1993	114	0.004	0.29	0.000	0.016	22,539	2.6	114	13.0
	1994	2,607	0.004	0.29	0.001	0.016	22,539	58.8	114	297.2
	1995-1999									
		125,161			**0.0455**			**3,599.5**		**14,268.4**

Table 5.1-3 ICPP Receipts and Constituent Quantities by Year

Period	Receipts (kg)	Pu (0.679 pCi/g U)		Np (1.82 pCi/g U)		Tc (154 pCi/g U)	
		ppb	g	ppb	g	ppm	g
1953	102	0.05	0.000005	2550	0.26	9	0.92
1954	231	0.05	0.000011	2550	0.59	9	2.08
1955	828	0.05	0.000040	2550	2.11	9	7.45
1956	744	0.05	0.000036	2550	1.90	9	6.70
1957	797	0.05	0.000038	2550	2.03	9	7.17
1958	898	0.05	0.000043	2550	2.29	9	8.08
1959	3741	0.05	0.000180	2550	9.54	9	33.67
1960	769	0.05	0.000037	2550	1.96	9	6.92
1961							
1962	775	0.05	0.000037	2550	1.98	9	6.98
1963							
1964	771	0.05	0.000037	2550	1.97	9	6.94
1965	425	0.05	0.000020	2550	1.08	9	3.83
1966	1408	0.05	0.000068	2550	3.59	9	12.67
1967							
1968	394	0.05	0.000019	2550	1.00	9	3.55
1969	427	0.05	0.000021	2550	1.09	9	3.84
1970	108	0.05	0.000005	2550	0.28	9	0.97
1971	1660	0.05	0.000080	2550	4.23	9	14.94
1972	413	0.05	0.000020	2550	1.05	9	3.72
1973	563	0.05	0.000027	2550	1.44	9	5.07
1974							
1975	1702	0.05	0.000082	2550	4.34	9	15.32
1976	195	0.05	0.000009	2550	0.50	9	1.76
1977	1333	0.05	0.000064	2550	3.40	9	12.00
1978	526	0.05	0.000025	2550	1.34	9	4.73
1979	535	0.05	0.000026	2550	1.36	9	4.82
1980	-1	0.05	0.000000	2550	0.00	9	-0.01
1981	905	0.05	0.000044	2550	2.31	9	8.15
1982	576	0.05	0.000028	2550	1.47	9	5.18
1983	1041	0.05	0.000050	2550	2.65	9	9.37
1984	2868	0.05	0.000138	2550	7.31	9	25.81
1985	1	0.05	0.000000	2550	0.00	9	0.01
1986	960	0.05	0.000046	2550	2.45	9	8.64
1987-1988							
1989	1	0.05	0.000000	2550	0.00	9	0.01
1990-1999							
	25,696		**0.001239**		**65.52**		**231.28**

NOTE: *Tables 5.1-2 and 5.1-3 report ppb values for Pu to two significant digits (X.XX), based upon the laboratory data available for this constituent. This raw data reported Pu mCi/g U to three decimal places, and this degree of reporting was maintained through the conversion to ppb. It should not be interpreted as accurate at the ppb level to two decimal places, but recognized as a product of the calculation.*

As an example, the average Pu activity for the base period was 0.004 μCi/g U. The representative Pu activity for each year (1964 through 1972) was calculated by multiplying 0.004 by 0.43/0.26, to yield 0.007 μCi/g U. This value was then assigned as the Pu activity for 1964 through 1972.

1955 through 1964

No data (constituent levels or alpha ratios) were located for SRS materials prior to December 1964. To arrive at values for this timeframe, data were extrapolated from the nearest period of time for which data were available (although limited), which was the period of December 1964 through May 1972. The Pu and Np activities from this adjacent timeframe (roughly 8 years) were applied to each year 1955 through 1964.

Recall that the Pu and Np constituent levels for the 1964-1972 period of time were based upon reported alpha ratios for those years, adjusted by the fraction of the alpha ratio for Pu and Np from the base period (1977 through 1984). As a result, the values extrapolated for 1953 through 1964 present a lesser degree of certainty than other values reported here.

It is also important to recognize that, although this period represents over one third of the time that RU materials were being shipped/processed at the Y-12 Complex, the corresponding amount of material is only slightly over 10% of the total material sent to the plant from SRS. These were the first years of the RU program and the initial shipments were substantially smaller than those that would follow once the program became fully operational.

Summary for Pu and Np Receipts

Chapter 4 of the SRS Site Report for RU flow notes that for Pu and Np "analytical results for uranium shipments from the site were available for only a small portion of the uranium shipped from SRS over the years." The report further provides a "most likely" concentration for Pu of 0.251 ppb and 73.4 ppb for Np.

The Np figure can be adjusted, based upon a uranium concentration in solution, to arrive at the ppb level for Np on a U basis. This concentration appears to be 6.82 gU/liter (based upon a four-year average from other sources). Making this adjustment for Np at a concentration of 116.5 dpm/ml yields a revised figure of 11,021 ppb. An appendix in the SRS Report offers another value for Np concentration of 242 dpm/ml. This figure results in Np levels of 25,739 ppb.

A comparison of the Y-12 Complex data shows good relative agreement with the SRS limited data for Pu. Np levels in the Y-12 Complex data, however, appear to be somewhat higher than those offered in the SRS report. Since the Y-12 Complex numbers are based upon a sizeable quantity of available data from the analytical laboratories over a period of several years, those values were used for this report.

Constituent quantities from the ICPP Site Report for Pu and Np[4] are significantly lower than those determined from the Y-12 Complex data for SRS material. SRS concentrations for Np average 29,221 ppb, while ICPP reports 2,550 ppb for their material. Additionally, SRS reports an average Pu concentration of 0.38 ppb, compared to ICPP's value of 0.05 ppb. Table 5.1-4 provides a summary of the quantities received at the Y-12 Complex from each site.

[4] ICPP, *Recycle Uranium Mass Balance Project, Idaho Site Report*, INEEL/INT-99-01228.

Table 5.1-4 Pu and Np Total Gram Quantities Received

	Plutonium g	Neptunium g
SRS	0.05	3,600
ICPP	0.0012	66
TOTALS	**0.051**	**3,666**

The reason for the differences in the respective constituent values for SRS versus ICPP is attributed to the different source of the material. ICPP reprocessed spent fuel from naval reactors and research reactors, while SRS reprocessed spent fuel from the production of plutonium. As a result, the spent fuel reprocessed at ICPP was "once through," i.e., not processed and subsequently recycled to the reactor. Accordingly, it would not have had the opportunity to accumulate the quantities of Pu, Np, and Tc that recycled material from multiple fuel cycles would have accumulated.

5.1.2.2 Shipments

Although data for shipments to SRS were not as readily available as that found for material receipts, a limited amount of information was located that addressed constituent levels in shipments for some of the time.

As with the RU receipts, a base period was established from which data was extrapolated to cover those years where data was otherwise not available. The base period for shipments was the time period of 1977 through 1984, with some additional data available for 1986 through 1989. Those years prior to 1977 (1953 through 1976) had no representative data that could be located. Also as with material receipts, the base period is discussed first.

1977 through 1984

Data for this timeframe was predominantly in the form of laboratory reports and/or logbook-type records maintained by the operating group(s). The latter was often a chronological listing of the individual lab results that were routinely received and recorded as a normal part of the operation.

Data from the various sources was generally found to be in good agreement, and the West presentation provided comparable results. Data typically included the date, material code/description, ID numbers, constituent activities, alpha ratio, beta ratio, and total uranium alpha. Typically, several individual analyses or data sets were available for each year. The parameters of interest from this raw data were averaged to arrive at representative values for each year and subsequently applied to material shipments to determine total quantities of Pu and Np.

1953 through 1976, 1985

Representative data for this large period of time could not be located for purposes of this report. Consequently, the average values for the Pu (0.004 μCi/g U) and Np (0.006 μCi/g U) activities from the years 1977 through 1984 (the shipment base period) were applied to each of these years.

While this extrapolation is more easily acceptable for years immediately adjacent to the base period, its application to materials shipped in years greatly separated from this period (i.e. the 1950s and early 1960s) has considerably less certainty. Nevertheless, no other data was located that represented these early years for materials shipped.

1986 through 1989

Metal button data were located that are representative of the product for this period of time. The respective activities are Pu 0.002 µCi/g U and Np 0.003 µCi/g U. Although the constituent activities differ somewhat from those reported for the base period, they were not used as a basis for calculating the quantities of these constituents.

Summary for Pu and Np Shipments

The SRS RU Site Report does not provide any quantitative constituent information for comparison to the limited data for shipments from the Y-12 Complex.

In determining the gram quantities of Pu and Np shipped from the plant, the respective activity levels in the shipped material are adjusted by the constituent specific activities to arrive at ppb. This number is then figured with gross material shipments to determine the total amounts, typically by year.

For reasons of classification, however, an annualized report of shipments to SRS cannot be provided in this document. Consequently, the base period activity levels are applied against the total quantity of material shipped (120,384 kg U) to generate the total grams of each constituent. These figures are provided in Table 5.1-5.

Table 5.1-5 Pu and Np Total Grams Shipped

Plutonium		Neptunium	
ppb	g	ppb	g
0.27	0.033	8,917	1,073

5.1.3 Technetium

5.1.3.1 Receipts

No data were found that provided quantitative information on the actual constituent level for Tc. Technetium levels were indirectly monitored through the use of the beta ratio for purposes of employee exposure control, and although this ratio was often reported, Tc was not the only constituent present in RU that contributed to the measured activity. (The ratio was defined as the ratio of beta radiation of the RU sample to the beta radiation for unirradiated uranium.)

Beta ratios were reported in analytical data for years 1977 through 1984, with an average value for all of those years equal to 0.97. Attributing all of this activity to Tc, the formula for the beta ratio can be solved to yield a Tc constituent level of 114 ppm. This does not appear to be unreasonable since many of the beta sources in unirradiated uranium have a relatively short half life, leaving Tc and americium (from Pu^{241} decay) as the predominant beta sources. Attributing all the activity to Tc is a conservative approach.

In the absence of Y-12 Complex qualitative data specific to Tc, the beta ratio is used as a basis for establishing Tc levels. This value is normally reported with the laboratory data for Pu and Np and provides a conservative basis for Tc levels.

The SRS Report cites 1983 as the only year for which an analysis of technetium in the uranium product stream was reported (DPST-84-385) and gives a typical concentration of 82 ppm. This figure is accompanied by a disclaimer that notes, “No claims are made as to the applicability of study results to other SRS production years.” In light of the uncertainty of the SRS value, this mass balance report was completed using a Tc concentration based upon the beta ratios reported in Y-12 Complex analytical reports over a period of several years and extrapolated to years not represented in those reports.

The ICPP report provides Tc levels (9 ppm) for the material that was sent to the Y-12 Complex from that facility. No Y-12 Complex data were located that were clearly identified as the ICPP material Tc levels, therefore the 9 ppm value was accepted for this material.

The technetium constituent quantities by year from Savannah River are included in Table 5.1-2, while Table 5.1-3 presents the quantities for Tc from the ICPP. Table 5.1-6 reports comparative ppm levels from SRS and ICPP, and the total grams received.

Table 5.1-6 Tc Total Grams Received

	Technetium	
	ppm	g
SRS	114	14,268
ICPP	9	231
TOTAL		**14,499**

5.1.3.2 Shipments

The SRS Site Report did not offer any information relative to the Tc concentration in metal received from the Y-12 Complex. In addition, very limited data was available at the Y-12 Complex that provided Tc concentrations, except for the years 1977 through 1980 and 1981. The Uranium Radioactivities Reports for these years did provide beta ratios for the materials shipped as metal to SRS.

The average beta ratio for those years was 0.87 with a range of only 0.82 to 0.90. If the same approach is taken for shipments that was discussed for receipts, i.e. attributing all beta activity to Tc, the average concentration is 102 ppm.

This approach was taken to report the Tc constituent levels that were shipped in the product to SRS and is reflected in Table 5.1-7. This provides a consistent methodology for receipts and shipments for this constituent and a more reasonable basis for comparison.

Table 5.1-7 Tc Total Grams Shipped

Technetium	
ppm	g
102	12,279

5.1.4 On-Site Inventory

The inventory still on-site at the Y-12 Complex (13 MTU) is primarily composed of metal-button product (9.7 MTU) and uranium-aluminum metal (U-Al) from SRS (3.3 MTU) that was not processed.

Button constituent quantities for Pu and Np were determined through the application of the Y-12 Complex data that reported activities of 0.002 μCi/g U (0.14 ppb) for Pu and 0.003 μCi/g U (4783 ppb) for Np for metal product from years circa 1986 -1989.

A technetium level of 102 ppm based upon a beta ratio wholly attributable to Tc (for materials shipped to SRS) yields the Tc concentration for this material.

The later year activity levels for incoming UN were applied to the U-Al material. These were based upon Y-12 Complex data for Pu and Np (0.004 μCi/g U and 0.016 μCi/g U, respectively). The Tc concentration was derived from the average beta ratio (114 ppm) for receipts from that same general timeframe. The results of the application of this data to the on-site inventory are provided in Table 5.1-8.

Table 5.1-8 Constituents in On-Site Inventory

	Pu		Np		Tc	
U-Al (3,300 kg)	0.29 ppb	0.001 g	22,539 ppb	74.4 g	114 ppm	376 g
Buttons (9,700 kg)	0.14 ppb	0.001 g	4,783 ppb	46.4 g	102 ppm	989 g
TOTALS		**0.002 g**		**120.8 g**		**1,365 g**

5.1.5 Waste

RU constituents were routinely discharged from the HEU processing complex in various effluent streams. As noted elsewhere in this report, the primary discharge point was the S-3 Ponds (until WETF became available in the mid-1980s). Other less significant discharge points include the Bear Creek Burial Grounds (for contaminated solid residues and process waste) and New Hope Pond (for surface contamination entrained by rainwater and secondary process wastewater).

The S-3 Pond sludge was extensively sampled and analyzed for metals and radioactive components prior to pond closure in the 1985-1986 timeframe. In a 1985 study of the exposure potential from S-3 Pond sludge,[5] C.M. West reported the results of radiological analysis for the sludge from each pond after neutralization, biodenitrification, and removal of the aqueous phase. The report provides the activity for the constituents of concern per gram of sludge (pCi/g wet weight). The average depth of the sludge for each pond is also provided. If the volume of sludge is approximated for each pond, the total volume can be applied to the activities from this report to arrive at the actual gram quantity of each constituent. Table 5.1-9 shows the results of these calculations.

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[5] Internal Correspondence, *Exposure Potential From S-3 Pond Dried Sludge*, C.M. West to H. D. Whitehead, Jr., June 3, 1985.

Table 5.1-9 Constituent Quantities in S-3 Pond Sludge

Pond	Activities (pCi)				
	Volume (ft^3)	^{238}Pu	$^{239, 240}Pu$	Np	Tc
SW	52,000	4.37E10	4.21E9	1.15E10	1.51E12
NW	84,000	3.93E10	<4.19E9	1.49E10	3.14E12
NE	124,000	1.20E10	8.89E9	4.64E10	3.05E12
SE	122,000	2.09E10	<6.08E9	3.04E10	4.56E13
	Total Curies	0.1159	*	0.1032	53.3
	Total Grams	0.0068	*	145	3,136

** The analysis of $^{239, 240}Pu$ was not statistically valid.*

If the SRS isotopic distribution for Pu is assumed (i.e., 84% ^{238}Pu, see Section 5.1.2), the total quantity of Pu in the pond sludge is estimated to be 0.008 g. The total amount of Tc is estimated at 3,136 g and Np at 145 g.

Residues and solids placed in the Bear Creek Burial Grounds were non-homogeneous and difficult to sample. Consequently, there are no RU exposure analyses or inventory numbers associated with the burial ground operation in the present analysis. However, since all solids leaving the HEU process area were extensively acid leached to recover residual uranium, potential impacts of RU constituents in Bear Creek are believed to be significantly less than in the S-3 Ponds. The Bear Creek waste management unit was also closed and capped in the same timeframe as the S-3 Ponds.

Contaminants collected in New Hope Pond were removed along with coal sediment from the Y-12 Complex coal storage yard on two different occasions (early 1970s and later 1980s) and placed in an unlined disposal basin on Chestnut Ridge above the water table. Fractional dose considerations associated with the routine operation and closure of New Hope Pond were judged to be less significant that similar activities around the S-3 Ponds. Like the S-3 Pond sludge, the New Hope Pond sediment was sampled and analyzed for radionuclides.

Disposal records and available analytical data for the West End Treatment Facility sludge storage tanks and New Hope Pond[6,7,8] were also reviewed. These records collectively indicated the presence of less than 0.01 g Pu, around 124 g of Np, and approximately 59 g Tc. Table 5.1-10 presents the waste figures in summary.

Table 5.1-10 Constituent Quantities Present in Waste Streams

Location	Pu (g)	Np (g)	Tc (g)
S-3 Ponds	0.008	145	3,136
WETF	<0.001	100	~50*
New Hope Pond	0.004	24	9
TOTALS	**~0.01**	**269**	**3,200**

**The Tc quantity for WETF was estimated based upon known process flows.*

[6] Saunders, M.B., *Leachability of Samples from New Hope Pond Disposal Basin,* Y/DZ-81, July 26, 1983.
[7] Internal Correspondence, "Transuranic Elements in Sediments from New Hope Pond and Sediment Basin," G.G. Fee to H.D. Hickman, March 16, 1984.
[8] Internal Correspondence, "Modified Surface Contamination Limits for WETF Sludge Project, G.R. Galloway to R.W. Oliver et al., August 12, 1997.

5.1.6 Mass Balance

The resulting mass balance for highly enriched RU and constituent flow through the Y-12 Complex is summarized in Table 5.1-11. This table compiles quantities of each constituent based upon the estimating logic presented in the preceding sections.

Table 5.1-11 Overall Mass Balance for Y-12 Complex Highly Enriched RU

	Receipts	Shipments	Inventory	Waste	Difference
RU (kg U)	150,857	120,384	13,082	~100	~17,300
Pu (g)	0.051	0.033	0.002	~0.01	~0
Np (g)	3,666	1,073	121	270	2,200 (-300)*
Tc (g)	14,499	12,279	1,365	3,200	-2,345 (335)†

** The Np difference is –300 g if it is assumed that the reported 1.75 Ci (2,500 g) Np was buried in the Bear Creek Burial Grounds as solid waste or shipped off site to another DOE facility.*
† The Tc difference is 340 g if it is assumed that most Tc found in the southeast S-3 Pond came from ORGDP and is not included in receipts.

Chapter 3 provided mass balance information at the RU level, reporting approximately 17.3 MT highly enriched RU that is not specifically accounted for. As explained previously, this difference is primarily attributable to the inability to precisely distinguish between RU and non-RU shipments.

Based upon Y-12 Complex records of highly enriched RU receipts and shipments, material remaining in inventory, and determinations regarding quantities in disposal, there remain no more than trace quantities of Pu not accounted for.

In contrast, the overall mass balance based primarily on receipt and shipment records cannot account for 2,200 g of Np. In the historical plant record,[9] reference is made to discharge of 2,500 g (1.75 Ci) of Np to the S-3 Ponds. As shown in Table 5.1-9, however, the amount of Np that can be accounted for by sampling and analysis of pond sludge is only 145 g. A similar quantity was found in the WETF sludge. It is known by a few individuals in the plant that an ion exchange column was installed in the uranyl nitrate feed stream to specifically remove Np from the incoming SRS RU for use in another program. The spent or loaded ion exchange columns were removed from the feed line and sent off-site for Np recovery. Since there was little residual uranium contained on the ion exchange resin, this transaction was not listed as an RU transfer and was not placed in the plant uranium accountability record. Assuming that the 2,500 g of Np identified in the waste management record was indeed separated from the RU stream as suspected and either sent off-site for use elsewhere or buried as a solid waste in the Bear Creek Burial Grounds, the overall mass balance shows 300 g more Np than can be accounted for.

Additional historical information was received from Y-12 Complex operations regarding Np recovery operations just prior to the issuance of this report. The information included a 1962 summary description of the Np recovery process for shipments of dilute uranyl nitrate from SRS,[10] communications of radiological protection safety measures,[11] and early years of analysis results for transuranics in SRS material.[12] Time did not permit further analysis for this report.

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[9] U.S. Department of Energy, *Historical Radionuclide Releases from Current DOE Oak Ridge Operations Office Facilities*, 1988.
[10] Internal Correspondence, "Np-237 Operations," R.E. Trent to J.R. Barkman, April 5,1962.

The overall Y-12 Complex mass balance shows 2,345 grams more Tc on the plant site than can be accounted for, based on the mass difference between the uranium feed, product, and waste streams. It should be noted that the normal flow of acid waste from the 9212 and 9206 HEU operations to the S-3 Ponds went first into the NE basin. The flow was then routed by overflow pipe to the NW basin, then to the SW basin, and finally into the SE basin. Under this normal design flow pattern, one would expect to find the greatest concentration of Tc in the NE basin and the least in the SE basin. Sludge analysis, however, shows 179 g of Tc in the NE basin, 184 g in the NW, 89 g in the SW, and 2,680 g in the SE. The apparent discrepancy was explained by a former S-3 Pond manager, who stated that on several occasions Tc liquid waste was discharged directly to the SE basin from 5-gal waste drums received from ORGDP. These Tc residues were removed from the gaseous diffusion cascade from time to time during certain maintenance activities. If it is assumed that essentially all of the Tc in the SE basin came from ORGDP and was not included in the Y-12 Complex RU database, the mass balance difference is 335 g Tc, or 2% of the estimated total receipt.

5.2 POTENTIAL AREAS OF CONCENTRATION

A steady state process model was developed for the HEU-process flow sheet to help identify the likely RU constituent accumulation points in the various units that make up the Y-12 Complex HEU chemical operations and to provide order-of-magnitude estimates of stream compositions for fractional dose calculations using the prescribed DOE methodology. The output of the HEU-process model for the Savannah River case is given in Appendix B; stream numbers correspond to the process block diagram numbers given in Chapter 2. Accountability records and combined SRS and Y-12 Complex analytical data were used to establish the feed stream compositions as the basis for the particular calculations shown. The overall results and conclusions of this assessment are driven largely by the unusually high concentration of ^{236}U in the SRS RU.

The SRS case shows the greatest potential for exposure of the Y-12 Complex population, subjugating the fractional impact of the ICPP RU. This bulk difference between the SRS RU and ICPP RU is explained by the higher level of ^{236}U in the SRS feed stream (27.8% ^{236}U average) compared to ICPP (<10% ^{236}U) and the fact that most of the RU processed at the Y-12 Complex was received from SRS (125 MTU) versus ICPP (26 MTU).

Most of the highly enriched RU material processed at the Y-12 Complex was in the form of fairly pure uranyl nitrate (UN) solution or uranium oxide (chiefly UO_2, UO_3, and/or U_3O_8). Smaller amounts of RU alloy (e.g., U-Al), casting dross, floor sweepings, and various residues were also received. Relatively pure alloys and oxides were first converted to UN solution, mixed with incoming UN solution, and then fed directly to the secondary solvent extraction system for concentration and purification. Two solvent extraction systems were used to purify the HEU; the first employing dibutyl carbitol as the extractor, and the second, tributyl phosphate (TBP). Neither organic purification process was capable of discriminating ^{236}U from ^{235}U or ^{238}U. Consequently, the uranium isotope distribution in the HEU feed was unaltered throughout the

[11] Internal Correspondence, "Safety Measures for Np-237 Processing," J.S. Reece to J.R. Barkman, September 9, 1960.
[12] Internal Correspondence, "Trans-Uranium Elements in SRO Material," R.H. Kent to J.R. Barkman, December 7, 1964.

HEU chemical facility. It should be noted that the Y-12 Complex solvent extraction systems were designed and operated specifically to remove elemental weapons system contaminants, such as C, Fe, and Cr, from UN with minimum loss of HEU to the acid waste raffinate stream. Further, the Y-12 Complex process was not modified specifically to remove TRU elements (i.e., Pu and Np) or various RU fission products (e.g., Tc) from the feed HEU. As a result, the RU components were allowed to distribute among the various process streams without design or specific process controls.

Available analytical data show that the majority of the radionuclides of interest tended to follow the uranium through the aqueous process and, consequently, largely ended up in the HEU metal buttons. It is estimated from the model that from 60 to 80% of the TRU components fed to the chemical process ended up in the HEU metal. The behavior of Tc is less certain since less analytical data was recorded for this RU constituent. However, based on beta-ratio data of the solvent extraction raffinate streams, it can be inferred that the bulk (i.e., >90%) of the Tc present in the process feed likely ended up in the HEU metal product.

The calculated results indicate that RU components moderately concentrated in the primary and secondary solvent extraction raffinate streams relative to the uranium flows. Analytical data taken from the S-3 Ponds and metal buttons are consistent with this model. The primary solvent extraction system raffinate was discharged to the S-3 Ponds during virtually all of the RU campaigns. The secondary system raffinate, on the other hand, was recycled to the primary system. Normally, one would expect the S-3 Ponds (stream 33, Appendix B) to be as concentrated in RU radioactivity as the primary raffinate. However, as shown in the process flow diagram in Appendix B, depleted uranium waste from other Y-12 Complex operations was also added to the S-3 Ponds (stream 31) and, later, to the WETF. The DU addition significantly diluted the TRU elements and fission products as well as the ^{236}U (since the DU contained little RU), making the S-3 Ponds and WETF less of a radiological hazard relative to unirradiated uranium.

5.3 POTENTIAL FOR WORKER EXPOSURE

Historically, worker protection from transuranics (^{238}Pu, ^{239}Pu, ^{240}Pu, and ^{237}Np) was provided by health physics programs for operators working with enriched, normal, or depleted uranium. This is due, in part, to the common method of detection (total alpha counting). Alpha particles emanating from uranium and transuranics are detectable in air and smear samples as well as urine samples collected routinely from uranium workers. As shown in Table 5.3-1, transuranics are significantly more active than even enriched uranium and therefore much more detectable.

A comprehensive health physics worker protection program, including radiation dosimetry, air sampling of the workplace, and routine urinalysis for identifying both uranium and total alpha level, was in place during the years that RU was processed at the Y-12 Complex. Health Physics Progress Reports were published on a semi-annual basis. Upon a limited review, it appears that the format and content of these reports remained relatively the same through the years. The progress report for the period January 1, 1953, to June 30, 1953, was selected for additional review because this reporting period coincides approximately with the time that RU was first processed at the Y-12 Complex and exemplifies the programs in place at that time.[13] The following information is summarized and excerpted from that report:

Table 5.3-1 Comparison of Specific Activity of Uranium and Transuranics

Material	Specific Activity (dpm per microgram)
Normal Uranium	1.5
^{234}U	13,764
^{235}U	5
^{236}U	144
^{237}Np	1,550
^{238}Pu	37,962,000
^{239}Pu	138,000
^{240}Pu	501,720

Air Sampling

Routine air sampling continued at all uranium, beryllium, and mercury handling operations. Special operations involving other contaminants were checked. Sampling for uranium included 10,940 general air samples and 1,266 operational/breathing-zone samples. Of these, 198 (1.8%) of the general air samples and 451 (35.6%) of the operational/breathing-zone samples were greater than the maximum permissible level (MPL) of 70 dpm per cubic meter. Efforts to reduce airborne levels were in progress. The installation of new hoods and improved housekeeping in the foundry area had dropped concentrations from near the MPL in 1951 to less than 20% of the MPL by the second quarter of 1953.

Routine Monitoring

A total of 379 persons were regularly issued one or more film meters. The processed film included 7,436 regular issued badges, 5,932 regular issued rings, 975 visitor badges, and 420 neutron film badges.

Routine Analysis

During the period January 1 – June 30, 1953, a total of 8,750 uranium analyses were made: 5,274 by the electroplating method and 3,476 by the fluorometric method. Most results for workers processing normal uranium were less than 20 μg per 24 hours. A small percentage (occurring only in weeks 2, 3, and 18) were between 20 and 30 μg per 24 hours. Urinalysis results for "enhanced" urinalysis (workers processing enriched uranium) ranged as high as 90 μg per 24 hours with approximately 10% of the results greater than 40 μg per 24 hours.

[13] Carbide and Carbon Chemicals Co., *Health Physics Progress Report*, January 1, 1953 through June 30, 1953, Union Carbide and Carbon Company, Oak Ridge, TN.

In addition, it was well known that transuranics as well as increased levels of ^{236}U were present in the RU received from SRS and ICPP. With this knowledge, specific analyses for transuranics were performed as appropriate, first by separation of the transuranics by chemical means and, after about 1960, by alpha spectroscopy techniques.

Beginning in 1961, about 2,200 employees per year were routinely monitored by bioassay and also in vivo techniques for internal exposure to uranium. In the period from 1961 to 1976, 49 employees were restricted from uranium work because they exceeded the established Plant Action Values (PAVs), a restriction rate of less than 0.05% per year of those being monitored.[14]

Worker Protection by Virtue of Specification

Prior to and during the processing of RU, the Y-12 Complex also operated as a uranium-processing facility. Careful consideration for worker protection was given to the introduction of RU for processing. A criterion for acceptance was based upon DOE/OR-859[15] which in turn, was derived from an informal agreement between the Y-12 Complex and SRS. The intent of this criterion was to maintain the relative hazard potential of all non-uranium alpha emitters to less than 7% of the relative hazard potential of uranium.[16] With this limitation, it was expected that RU could be safely managed by the measures already in place for processing uranium. The specification for RU included a limit for alpha activity in the form of the alpha ratio and also a limit on the level of gamma and beta activity. A detailed explanation of these specifications may be found in Section 4.3.

In the 1985 presentation to DOE compiled by Y-12 Complex Health Physicist, C.M. West, [17] reactor returns (RU) were considered for the period of 1953 to 1984. Not only were incoming levels of transuranics allowed by specification investigated, but also any levels concentrated by processing at the Y-12 Complex.

A study to evaluate worker average exposure when working with RU was conducted at the Y-12 Complex from 1980 to 1984, comparing operators working with RU to workers in the same department not working with RU. The results are shown in Table 5.3-2.

This difference is considered the upper level of exposure due to the processing of reactor returns. The presentation included the conclusion that exposures at this level were not considered to be a significant health risk.

Average results from general air samples (60,000) taken in areas where RU was processed from 1977 to 1985 were 3% of the uranium radioactivity concentration standard. The average alpha ratio for RU for these years was 30% of the specification. This specification was set to control exposure from plutonium to 7% of that from uranium. Using this data, the estimated internal dose (committed dose to bone) from transuranics was calculated to be 0.019 rem per year, considered to be an acceptable health risk.

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[14] West, C.M., et al., *Sixteen Years of Uranium Personnel Monitoring Experience in Retrospect*, Union Carbide Company, July 1977.
[15] Egli et al., *The Report of the Joint Task Force on Uranium Recycle Materials Processing*, 1985.
[16] Vath and Duerksen, *Criteria for Acceptance and Technical Assessment for Acceptance of Enriched Uranium at the Y-12 Plant*, April 25, 1996.
[17] West, C.M., *Radioactive Contaminants in Uranium Reactor Returns at the Oak Ridge Plant*, 1985.

Table 5.3-2 Comparison of Exposures of Workers Handling Reactor Returns to Others in Same Department Not Handling Reactor Returns. Five-Year (1980 – 1984) Average Exposures in rem/year.

Group	Average No. of Workers/Year	Skin Exposure (rem/year)	Penetrating Exposure (rem/year)
Working with reactor returns	22	0.524	0.305
Others in same department not working with reactor returns	180	0.176	0.112
DIFFERENCE		0.348	0.193

5.4 POTENTIAL FOR ENVIRONMENTAL CONTAMINATION

As presented in Section 2.5, environmental monitoring has been performed on- and off-site at the Y-12 Complex since about 1953. As could be expected from the plant operating history, the most significant material releases have been uranium. Table 5.4-1 below presents a summary of radionuclide releases from the Y-12 Complex.[18]

Table 5.4-1 Summary of Radionuclides Released to Air and Water or Buried at Y-12 Complex from 1944 through 1987

Radionuclide	Air (Curies)	Water (Curies)	Burial (Curies)*
Uranium	13.87 (6296 kg)	116.58 (182,374 kg)	7,097 (17,290,523 kg)
Thorium	-	0.680	18.59
Technetium			58.60†

* *Prior to 1972, liquid wastes that were transferred to the S-3 Ponds were recorded as burials.*
† *Approximately 2,680 grams received from ORGDP was recorded as a burial.*

The original table listed several radionuclides other than uranium, thorium, and technetium, including Np and Pu. These radionuclides (along with the Tc) were associated with recycled reactor product uranium solutions received from other DOE sites (primarily SRS and ICPP) since 1953. The recovery process for this solution resulted in some of these radionuclides remaining in the product (which was subsequently returned to SRS as metal buttons). The waste from the process went to the S-3 Ponds prior to about 1984 and was recorded as a burial. Since measurements at that time were made for contamination control purposes only, the exact quantities of these radionuclides that went to the ponds are unknown. Reporting thresholds were established for these radionuclides for accountability and security purposes. Releases of Np and Pu to the ponds were always below the reporting thresholds of 1.7 Ci and 0.87 Ci, respectively.

A joint task force was assembled by DOE in 1985 to study past and then-current practices relating to the processing of uranium recycle materials. From the data reviewed, the task force did not disclose any instance in which the environment, safety, or health of plant workers or the

[18] U.S. Department of Energy, *Historical Radionuclide Releases from Current Oak Ridge Operations Office Facilities*, 1988.

public were jeopardized or compromised. The primary recommendation was for the gaseous diffusion plants to develop formal specifications on maximum permissible levels of contaminants in enrichment feed materials. No recommendations were suggested regarding the releases from the Y-12 Complex.[19]

The Task 7 component of the Oak Ridge Dose Reconstruction Project, initiated in 1994, involved performing qualitative and quantitative screening of various materials of concern at the DOE Oak Ridge sites. Materials screened included Np and Tc. Based on the analysis of the data, the Task 7 team determined that Np did not warrant further study. Although Tc was identified as one of the potential candidates for further study, it was not determined to be a high-priority candidate.[20]

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[19] Egli et al., *Report of the Joint Task Force on Uranium Recycle Materials Processing*, 1985.
[20] Bruce, *Screening-Level Evaluation of Additional Potential Materials of Concern,* July 1999.